CAN BAC'N PCT/PTO 0 6 FEB 2002 FORM PTO-1390 (REV. 12-2001) U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE ATTORNEY 'S DOCKET NUMBER 1888 TRANSMITTAL LETTER TO THE UNITED STATES U.S. APPLICATION NO (If known, see 37 CFR 1.5 DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371 INTERNATIONAL APPLICATION NO. INTERNATIONAL FILING DATE 10 August 1999 (10.08.99) PCT/US00/21874 10 August 2000 (10.08.00) TITLE OF INVENTION COSMETICS APPLICANT(S) FOR DO/EO/US NATIONAL STARCH AND CHEMICAL INVESTMENT HOLDING CORPORATION Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information: 1. This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371. 2. This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371. This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below. 4. The US has been elected by the expiration of 19 months from the priority date (Article 31). 5. X A copy of the International Application as filed (35 U.S.C. 371(c)(2)) is attached hereto (required only if not communicated by the International Bureau). has been communicated by the International Bureau. is not required, as the application was filed in the United States Receiving Office (RO/US). €. An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)). is attached hereto. has been previously submitted under 35 U.S.C. 154(d)(4). 7. Amendments to the claims of the International Aplication under PCT Article 19 (35 U.S.C. 371(c)(3)) are attached hereto (required only if not communicated by the International Bureau). have been communicated by the International Bureau. have not been made; however, the time limit for making such amendments has NOT expired. d. Y have not been made and will not be made. 8. An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371 (c)(3)). 9. An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). 10. An English lanugage translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

#### Items 11 to 20 below concern document(s) or information included:

11.	An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. 🔀	An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13.X	A FIRST preliminary amendment.
14.	A SECOND or SUBSEQUENT preliminary amendment.
15.	A substitute specification.
16.	A change of power of attorney and/or address letter.
17.	A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
18.	A second copy of the published international application under 35 U.S.C. 154(d)(4).
19.	A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
20 🖂	Other stems or information:

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NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.								
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Bridgewater, New Jersey 08807-0500 REGISTRATION NUMBER								

**CASE 1888** 

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF

Group Art Unit:

TSUZUKI, ET AL.

Examiner:

S.N.

FILED: CONCURRENTLY HEREWITH

FOR: COSMETICS

Commissioner of Patents and Trademarks Washington, D.C. 20231

# PRELIMINARY AMENDMENT

Sir:

In the above-identified application, Applicants respectfully request the following preliminary amendment be entered and the claims considered in light thereof.

## IN THE CLAIMS

Cancel claims 1-13 and insert new claims 14-27 to read:

- 14. A cosmetic comprising an amphoteric urethane resin having at least one carboxyl group and at least one tertiary amino group in a molecule, and a silicone polymer.
- 15. The cosmetic according to claim 14, wherein said silicone polymer is a nonionic silicone polymer.
- 16. The cosmetic according to claim 14, wherein said silicone polymer is a polyether-modified silicone polymer.
- 17. The cosmetic according to claim 14, wherein said silicone polymer is a phenyl-modified silicone polymer.
- 18. The cosmetic according to claim 14, wherein said silicone polymer is an amino-modified silicone polymer.
- 19. The cosmetic according to claim 14, wherein said silicone polymer is an alkyl-modified silicone polymer.
- 20. The cosmetic according to claim 14, wherein said silicone polymer is an alkoxy-modified silicone polymer.
- 21. The cosmetic according to claim 14, wherein said silicone polymer is a cyclic silicone polymer.
- 22. The cosmetic according to claim 14, wherein said silicone polymer is in the form of an aqueous solution.
- 23. The cosmetic according to claim 14, wherein said silicone polymer is in the form of a water dispersion.
- 24. The cosmetic according to claim 14, wherein said amphoteric urethane resin has at least one structural unit derived from ethylene oxide in its structure.
- 25. The cosmetic according to claim 14, wherein said amphoteric urethane resin has at least one polysiloxane bond in its structure.
- 26. The cosmetic according to claim 14, wherein said amphoteric urethane resin is in the form of an aqueous solution.

27. The cosmetic according to claim 14, which is selected from the group consisting of a hair cosmetic, a skin care cosmetic and a make-up cosmetic.

# STATUS OF THE CLAIMS

Claims 1-13 were filed as PCT/US00/21874 internationally filed 10 August, 2000.

Claims 1-13 have been cancelled.

Claims 14-27 have been added. A marked up copy of the claims as amended is attached as Appendix A.

Claims 14-27 are presented for consideration. A clean copy of the claims as currently pending is attached as Appendix B.

## **REMARKS**

Claims 1-13 have been cancelled and new claims 14-27 have been added. Claims 14-27 have descriptive basis in the claims as filed in PCT/US00/21874, but have been amended to conform to standard US patent practice.

In view of the foregoing, Applicant respectfully requests early action in this application.

Respectfully submitted,

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# Appendix A (marked up copy of amended claims)

- [ 1. A cosmetic comprising an amphoteric urethane resin having carboxyl group(s) and tertiary amino group(s) in a molecule, and silicone polymer.
- 2. The cosmetic according to claim 1, wherein said silicone polymer is a nonionic silicone polymer.
- 3. The cosmetic according to claim 1, wherein said silicone polymer is a polyether-modified silicone polymer.
- 4. The cosmetic according to claim 1, wherein said silicone polymer is a phenyl-modified silicone polymer.
- 5. The cosmetic according to claim 1, wherein said silicone polymer is an amino-modified silicone polymer.
- 6. The cosmetic according to claim 1, wherein said silicone polymer is an alkyl-modified silicone polymer.
- 7. The cosmetic according to claim 1, wherein said silicone polymer is an alkoxy-modified silicone polymer.
- 8. The cosmetic according to claim 1, wherein said silicone polymer is a cyclic silicone polymer.
- 9. The cosmetic according to any one of claims 1 to 8, wherein said silicone polymer is in the form of an aqueous solution or a water dispersion.
- 10. The cosmetic according to any one of claims 1 to 9, wherein said amphoteric urethane resin has structural unit(s) derived from ethylene oxide in its structure.
- 11. The cosmetic according to any one of claims 1 to 9, wherein said amphoteric urethane resin has polysiloxane bond(s) in its structure.
- 12. The cosmetic according to any one of claims 1 to 11, wherein said amphoteric urethane resin is in the form of an aqueous solution.
- 13. The cosmetic according to any one of claims 1 to 12, which is at least one selected from the group consisting of hair cosmetics, skin care cosmetics and make-up cosmetics.]
- 14. A cosmetic comprising an amphoteric urethane resin having at least one carboxyl group and at least one tertiary amino group in a molecule, and a silicone polymer.
- 15. The cosmetic according to claim 14, wherein said silicone polymer is a nonionic silicone polymer.
- 16. The cosmetic according to claim 14, wherein said silicone polymer is a polyether-modified silicone polymer.
- 17. The cosmetic according to claim 14, wherein said silicone polymer is a phenyl-modified silicone polymer.

18. The cosmetic according to claim 14, wherein said silicone polymer is
an amino-modified silicone polymer.
19. The cosmetic according to claim 14, wherein said silicone polymer is
an alkyl-modified silicone polymer.
20. The cosmetic according to claim 14, wherein said silicone polymer is
an alkoxy-modified silicone polymer.
21. The cosmetic according to claim 14, wherein said silicone polymer is a
cyclic silicone polymer.
22. The cosmetic according to claim 14, wherein said silicone polymer is
in the form of an aqueous solution.
23. The cosmetic according to claim 14, wherein said silicone polymer is
in the form of a water dispersion.
24. The cosmetic according to claim 14, wherein said amphoteric urethane
resin has at least one structural unit derived from ethylene oxide in its structure.
25. The cosmetic according to claim 14, wherein said amphoteric urethane
resin has at least one polysiloxane bond in its structure.
26. The cosmetic according to claim 14, wherein said amphoteric urethane
resin is in the form of an aqueous solution.
27. The cosmetic according to claim 14, which is selected from the
group consisting of a hair cosmetic, a skin care cosmetic and a make-up
cosmetic.

# Appendix B (clean copy of pending claims)

- 14. A cosmetic comprising an amphoteric urethane resin having at least one carboxyl group and at least one tertiary amino group in a molecule, and a silicone polymer.
- 15. The cosmetic according to claim 14, wherein said silicone polymer is a nonionic silicone polymer.
- 16. The cosmetic according to claim 14, wherein said silicone polymer is a polyether-modified silicone polymer.
- 17. The cosmetic according to claim 14, wherein said silicone polymer is a phenyl-modified silicone polymer.
- 18. The cosmetic according to claim 14, wherein said silicone polymer is an amino-modified silicone polymer.
- 19. The cosmetic according to claim 14, wherein said silicone polymer is an alkyl-modified silicone polymer.
- 20. The cosmetic according to claim 14, wherein said silicone polymer is an alkoxy-modified silicone polymer.
- 21. The cosmetic according to claim 14, wherein said silicone polymer is a cyclic silicone polymer.
- 22. The cosmetic according to claim 14, wherein said silicone polymer is in the form of an aqueous solution.
- 23. The cosmetic according to claim 14, wherein said silicone polymer is in the form of a water dispersion.
- 24. The cosmetic according to claim 14, wherein said amphoteric urethane resin has at least one structural unit derived from ethylene oxide in its structure.
- 25. The cosmetic according to claim 14, wherein said amphoteric urethane resin has at least one polysiloxane bond in its structure.
- 26. The cosmetic according to claim 14, wherein said amphoteric urethane resin is in the form of an aqueous solution.
- 27. The cosmetic according to claim 14, which is selected from the group consisting of a hair cosmetic, a skin care cosmetic and a make-up cosmetic.

#### COSMETICS

#### Field of the Invention

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The present invention relates to cosmetics containing an amphoteric urethane resin and a silicone polymer.

#### Description of the Related Art

As a base resin for cosmetics such as hair fixatives, for example, water-soluble resins such as nonionic (non-ionic) polyvinyl pyrrolidone (PVP), cationic acrylic resin or cellulose, anionic acrylic resin or polyvinyl acetate, and amphoteric acrylic resin or polyvinyl acetate have hitherto been used. Hair fixatives comprising these water-soluble resins as the base resin have merits such as high curl holding power and excellent durability, but have problems such as poor touch, which is important for cosmetics, and drastically poor feel.

On the other hand, the present inventors found that an amphoteric urethane resin having carboxyl group(s) and tertiary amino group(s) in a molecule can provide excellent feel because of the elasticity and strong toughness of the urethane resin and filed a patent application with respect to a resin composition for cosmetics, comprising the amphoteric urethane resin as

the base resin (Japanese Patent Application No. Hei 10-27595).

However, the cosmetic comprising the amphoteric urethane resin as the base resin is superior in feel, but has such a problem that a friction coefficient of the surface is large and spread at the time of application is inferior because of poor surface smoothness.

#### Summary of the Invention

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The present invention has been accomplished under these circumstances and an object thereof is to provide a cosmetic which is superior in both characteristics of touch and spread at the time of application.

To attain the object described above, the present invention is directed to a cosmetic comprising an amphoteric urethane resin having carboxyl group(s) and tertiary amino group(s) in a molecule, and a silicone polymer.

The present inventors have studied intensively to solve the problems such as surface smoothness, which is a weak point in case of using the amphoteric urethane resin. As a result, they have found that, when using an amphoteric urethane resin in combination with a silicone polymer, good results are obtained. That is, since the

compatibility of the said amphoteric urethane resin with the silicone polymer is not high and the silicone polymer has stronger hydrophobicity, the amphoteric urethane resin and silicone polymer cause micro phase separation and the silicone polymer is unevenly distributed on the surface, thereby making it possible to provide the surface with the smoothness. As a result, they have found that a cosmetic comprising the amphoteric urethane resin in combination with the silicone polymer is superior in spread at the time of application without impairing the touch when using the amphoteric urethane resin alone, thus completing the present invention.

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When using an aqueous solution or a water dispersion of the silicone polymer as the silicone polymer, the compatibility with the amphoteric urethane resin is enhanced to some degree, thereby making it possible to prepare a cosmetic having good stability.

When a structural unit derived from ethylene oxide is introduced, as a nonionic hydrophilic component, into the structure of the amphoteric urethane resin, sufficient hydrophilicity can be obtained and the hair washability is particularly improved when using as a hair cosmetic.

By. introducing polysiloxane bond(s) into the structure of the said amphoteric urethane resin, the touch is further improved when using as a hair cosmetic.

# Description of the Preferred Embodiments

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The embodiments of the present invention will now be described below.

The cosmetics of the present invention can be obtained by using an amphoteric urethane resin having carboxyl group(s) and tertiary amino group(s) in a molecule, and a silicone polymer.

The cosmetics of the present invention are used as hair cosmetics such as foam hair fixative, gel-like hair fixative, aerosol spray hair fixative, pump spray hair fixative and creamy hair fixative; skin care cosmetics such as shaving cream, skin care lotion and sunscreen lotion; and make-up cosmetics such as nail polish, mascara and foundation; and are particularly preferably used as hair cosmetics.

The amphoteric urethane resin having carboxyl group(s) and tertiary amino group(s) in a molecule can be prepared, for example, by reacting a polyol compound (component A), a polyisocyanate compound (component B) and a compound having active hydrogen(s) and carboxyl group(s) (component C) in the presence of excess isocyanate groups to form an isocyanate groupcontaining prepolymer, and reacting the isocyanate group-containing prepolymer with a compound having active hydrogen(s) and tertiary amino group(s)

(component D). Alternatively, the amphoteric urethane resin can also be prepared by replacing the sequence of the reaction between the above specific components C and D, that is, by reacting the above components A. B. and D in the presence of excess isocyanate groups to form an isocyanate group-containing prepolymer, and reacting the isocyanate group-containing prepolymer with the above specific component C. Such a method makes it possible to prepare an amphoteric urethane resin simply and safely as compared with a conventional method. In the above preparation method, when both specific components C and D are simultaneously reacted, together with the components A and B, the carboxyl group(s) in the component C and the tertiary amino group(s) in the component D form(s) a salt, which is insoluble in the reaction system. As a result, the reaction with the isocyanate compound does not occur even in the presence of the OH group(s), thereby making it impossible to prepare a desired amphoteric urethane resin.

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The above polyol compound (component A) is not specifically limited as far as it can be used in the preparation of a normal polyurethane, and examples thereof include polyester poylol, polyether polyol, polycarbonate polyol, polybutadiene polyol, polyisoprene

polyol, polyolefin polyol and polyacrylate polyol, etc.

These polyol compounds are used alone or in combination. Among these polyol compounds, polyester poylol and polyether polyol are preferably used.

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Examples of the polyester polyol include those obtained by polycondensing at least one of dicarboxylic acids such as succinic acid, glutaric acid, adipic acid, sebacic acid, azelaic acid, maleic acid, fumaric acid, phthalic acid and terephthalic acid with at least one of polyhydric alcohols such as ethylene glyol, propylene glycol, 1,4-butanediol, 1,3-butanediol, 1,6-hexanediol, neopentyl glycol, 1,8-octanediol, 1,10-decanediol. diethylene glycol, spiro-glycol and trimethylolpropane, etc., and those obtained bу the ring-opening polymerization of lactones.

Example of the polyether polyol include polyhydric alcohols used in the synthesis of the said polyester polyols, phenols such as bisphenol A, or those obtained by the ring-opening addition polymerization of primary amines or secondary amines and cyclic ether such as ethylene oxide, propylene oxide, oxetane and tetrahydrofuran. Examples thereof ınclude polyoxyethylene polyol, polyoxypropylene polyol. polyoxytetramethylene polyol, and those obtained by the ring-opening addition polymerization of bisphenol A and at least one of propylene oxide and ethylene oxide, etc.

(in case of a copolymer it may be either a block copolymer or a random copolymer).

The polyisocyanate compound (component B) is not specifically limited, and examples thereof include organic diisocyanate compounds such as aliphatic diisocyanate compound, alicyclic diisocyanate compound and aromatic diisocyanate compound. These compounds may be used alone or in combination.

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Example of the aliphatic diisocyanate compound includes ethylene diisocyanate, 2,2,4-trimethylhexamethylene diisocyanate, 1,6-hexamethylene diisocyanate, Examples of the alicyclic diisocyanate compound 4,4'-diphenylmethane includes hydrogenated diisocyanate, 1,4-cyclohexane diisocyanate, methylcyclohexylene diisocyanate, isophorone diisocyanate and norbornane diisocyanate, etc. Examples of the aromatic diisocyanate compound includes 4,4'-diphenylmethane diisocyanate, xylylene diisocyanate, toluene diisocyanate and naphthalene diisocyanate, etc. Among these compounds, 1,6hexamethylene diisocyanate, isophorone diisocyanate, norbornane diisocyanate, etc. are preferable because of the excellent light resistance and low price.

25 The compound (component C) having active

hydrogen(s) and carboxyl group(s) is not specifically limited as far as it is a compound having at least one active hydrogen and at least one carboxyl group in a molecule, and examples thereof include dimethylolpropionic acid (DMPA), dimethylolbutanoic acid, carboxyl group-containing polycaprolactone diol, etc. These compounds may be used alone or in combination.

The compound (component D) having the above active hydrogen(s) and tertiary amino group(s) is not specifically limited as far as it is a compound having at least one active hydrogen and at least one tertiary amino group in a molecule, and examples thereof include N-alkyldialkanolamine compound such as N-methyldiethanolamine and N-butyldiethanolamine, and dimethylaminoethanol, etc. These compounds may be used alone or in combination.

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In the production of an isocyanate groupcontaining prepolymer by using the above respective
components, chain extenders or molecular weight
inhibitors can be used for the purpose of controlling
various characteristics of the amphoteric urethane resin
as a final product.

The chain extender is not specifically limited and 25 examples thereof include low-molecular polyol, amines.

Examples of the low-molecular polyol includes etc. glycols such as ethylene glycol, propylene glycol, 1,4butanediol, diethylene glycol, 1,6-hexanediol, spiroglycol, cyclohexyl dimethanol, hydrogenated bisphenol A, neopentyl glycol, bis(beta-hydroxyethoxy)benzene, and xylylene glycol; and triol such as trimethylolpropane and glycerin. Examples of the amines include ethylenediamine, propylenediamine, piperazine, hydrazine, isophoronediamine, methylene(bis-ochloroaniline) and propylene glycol having amino groups at both ends, etc.

Examples of the molecular weight inhibitor includes propylene glycol having an amino group at one end, etc.

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In the production of the amphoteric urethane resin, solvents can be used as needed. For example, organic solvents capable of dissolving both raw materials and polyurethane to be prepared are preferably used. Examples of the organic solvent includes amides such as N-methylpyrrolidone, dimethylformamide and dimethylacetamide; ketones such as acetone and methyl ethyl ketone; esters such as ethyl acetate; cellosolve acetate, cellosolve ether, etc..

In the production of the amphoteric urethane resin, the dispersibility in water can be provided by

neutralizing the carboxyl group(s) or tertiary amino group(s) incorporated into the molecule with a neutralizing agent. Examples of the neutralizing agent for the said carboxyl group(s) includes triethylamine, trimethylamine, 2-amino-2-methyl-1-propanol, triethanolamine, potassium hydroxide, sodium hydroxide, etc. Examples of the nutralizing agent for the said tertiary amino group(s) include acetic acid, hydrochloric acid, sulfuric, nitric acid and dimethylsulfuric acid, etc.

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In the production of the amphoteric urethane resin, polymerization catalysts known in the field of polyurethane can be used, and examples thereof include tertiary amine catalyst, organometallic catalyst, etc. Examples of the tertiary amine catalyst includes [2,2,2]diazabicyclooctane (DABCO), tetramethyleneiamine, N-methyl morphorine and diazabicycloundecene (DBU), etc. Examples of the organometallic catalyst includes dibuty!tin di!aurate, etc.

As the amphoteric urethane resin, in view of the hair washability, those having structural unit(s) derived from ethylene oxide (EO) in the structure are preferably used.

Examples of the structural unit derived from the said EO includes EO unit represented by the general formula (I) described below, propylene oxide (hereinafter

abbreviated to "PO") unit represented by the general formula (II) described below, etc., and the EO unit is preferably used.

$$(CH2 CH2 O) ... (1)$$

$$\begin{array}{c} \text{CH}_3 \\ \hline - \left( \text{CHCH}_2 \text{ O} \right) \\ \hline \end{array} \qquad \dots \left( \text{II} \right)$$

The above amphoteric urethane resin may have both EO and PO units. A proportion of the EO unit to the PO unit is preferably within a range from 10/0 to 2/8, and particularly preferably from 10/0 to 4/6, on a weight basis.

The repeating number n of the EO unit in the general formula (I) is preferably set within a range from 3 to 300, and particularly preferably from 20 to 120. When the n is less than 3, sufficient hydrophilicity can not be provided because of too small amount of the EO unit(s) to be incorporated into the amphoteric urethane resin and, therefore, sufficient hair washability are not likely to be obtained. On the other hand, when n exceeds 300, an adverse influence is likely to be exerted on the moisture resistance or the like because of too strong hydrophilicity of the amphoteric urethane resin. Furthermore, the repeating number m of the PO unit in the general formula

(II) is preferably set within a range from 3 to 300, and particularly preferably from 20 to 120. When the amphoteric urethane resin has both EO and PO units, (n + m) is preferably set within a range from 3 to 300, and particularly preferably from 20 to 120.

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The amphoteric urethane resin having structural unit(s) derived from ethylene oxide (EO) can be prepared, for example, by reacting a polyol compound (component A), a polyisocyanate compound (component B), a polyethylene oxide derivatives having active hydrogen(s), and a compound having active hydrogen(s) and carboxyl group(s) (component C) in the presence of excess isocyanate groups to form an isocyanate groupcontaining prepolymer, and reacting the isocyanate group-containing prepolymer with a compound having active hydrogen(s) and tertiary amino group(s) (component D). Alternatively, the amphoteric urethane resin can also be prepared by replacing the sequence of the reaction between the above components C and D. As the above components A to D, the same compounds as those described above can be used.

Examples of the polyethylene oxide derivative having active hydrogen(s) to be used together with the above components A to D is not specifically limited as far as it is capable of introducing a structural unit derived

from ethylene oxide (EO) into the structure of the above amphoteric urethane resin, and examples thereof include polyoxyethylene glycol (PEG), polyoxyethylene polyoxypropylene glycol (EOPO block copolymer), etc. Among these, polyoxyethylene glycol is preferably used. The above polyoxyethylene oxide derivative may be any of a type wherein an OH group is introduced at both ends, a type wherein an NH2 group is introduced at both ends, a type wherein an OH group is introduced at one end, and a type wherein an NH2 group is introduced at one end. When using the type wherein an OH group is introduced at both ends or a type wherein an NH2 group is introduced at both ends, an amphoteric urethane resin having the EO unit(s) in a principal chain is obtained. When using the type wherein an OH group is introduced at one end or a type wherein an NH2 group is introduced at one end, an amphoteric urethane resin having EO unit(s) at its side chain(s) or end(s) is obtained.

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The molecular weight of the specific polyethylene oxide derivative is preferably within a range from 200 to 20000, and particularly preferably from 1000 to 10000

To further improve the feel, those having polysiloxane bond(s) in the structure are preferably used as the amphoteric urethane resin.

25 The repeating number n of a siloxane bond (Si-O)

is preferably within a range from 5 to 300, and particularly preferably from 20 to 150. When n is less than 5, it becomes difficult to obtain the sufficient effect on the touch, coming properties, etc. obtained intrinsically by introducing the polysiloxane bond(s) because of too small amount of the polysiloxane bond(s) in the resulting amphoteric urethane resin. On the other hand, when n exceeds 300, the compatibility with other raw materials become inferior because of high hydrophobicity, thereby making it difficult to react them. Furthermore, the adhesion to hair is likely to be inhibited because of too high hydrophobicity of the resulting polymer.

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The amphoteric urethane resin having the said polysiloxane bond(s) can be prepared, for example, by reacting a polyol compound (component A), a polyisocyanate compound (component B), a polysiloxane compound having active hydrogen(s) and a compound having active hydrogen(s), and carboxyl group(s) (component C) in the presence of excess isocyanate groups to form an isocyanate group-containing prepolymer, and reacting the isocyanate group-containing prepolymer with a compound having active hydrogen(s) and tertiary amino group(s) (component D). Alternatively, the amphoteric urethane resin can also be

prepared by replacing the sequence of the reaction between the above components C and D. As the above components A to D, the same compounds as those described above can be used.

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The polysilexane compound having active hydrogen(s) to be used together with the above components A to D is not specifically limited as far as it is capable of introducing polysiloxane bond(s) into the structure of the amphoteric urethane resin, and examples thereof polydialkylsiloxanediol, polydialkylinclude siloxanemonool, polydialkylsiloxanediamine, polydialkylsiloxanemonoamine, etc. These compounds may be used alone or in combination. The alkyl group(s) to be combined with Si of the respective siloxane bonds of the said polydialkylsiloxanediol preferably has 1 to 10 carbon atoms, and particularly preferably 1 to 5 carbon atoms. The above polysiloxane compound may contain various siloxane bonds wherein the number of carbon atoms of the alkyl group(s) to be combined with Si of the siloxane bonds varies. Specific examples of the polydialkylsiloxanediol include polydimethylsiloxanediol, polymethylethylsiloxanediol, etc. Examples of the polydialkylsiloxanemonool include polydimethyl-siloxanemonool, polymethylethyl-siloxanemonool, etc. Examples of the polydialkylsiloxanediamine include polydimethylsiloxanediamine, polymethylethyl-siloxanediamine, etc.

Examples of the polydialkyl-siloxanemonoamine include poly-dimethylsiloxane-monoamine,

polymethylethylsiloxane-monomaine, etc.

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type wherein an OH group is introduced at both ends, a type wherein an NH<sub>2</sub> group is introduced at both ends, a type wherein an OH group is introduced at one end and a type wherein an NH<sub>2</sub> group is introduced at one end. When using the type wherein an OH group is introduced at both ends or a type wherein an NH<sub>2</sub> group is introduced at both ends or a type wherein an NH<sub>2</sub> group is introduced at both ends, an amphoteric urethane resin having polysiloxane bond(s) in a principal chain is obtained. When using the type wherein an OH group is introduced at one end or a type wherein an NH<sub>2</sub> group is introduced at one end, an amphoteric urethane resin having polysiloxane bond(s) at its side chain(s) or end(s) is obtained.

In the cosmetics of the present invention, the amphoteric urethane resin is preferably used in the form of an aqueous solution. In the present invention, the aqueous solution includes not only an aqueous solution state where the amphoteric urethane resin is completely dissolved in water but also a water dispersion state

where the amphoteric urethane resin is dispersed in water.

It is also possible to add a crosslinking agent such as silane coupling agent to the water dispersion of the above amphoteric resin, thereby to provide the crosslinkability. To provide the storage stability, various additives may be added and examples thereof include protective colloidal agents, bactericides, mildewproofing agents, etc.

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The silicone polymer used together with the said amphoteric urethane resin is not specifically limited as far as it has polysiloxane bond(s) in a molecule and can be used in cosmetics, and examples thereof include silicone resin, silicone oil, silicone emulsion, silicone rubber, etc. Examples of the silicone rubber include nonionic, polyether-modified, phenyl-modified, aminomodified, alkyl-modified, alkoxy-modified, cyclic silicone polymers, etc. These silicone polymers can be used alone or in combination.

20 Examples of the nonionic silicone polymer includes those represented by the following general formula (1):

$$\begin{array}{c}
R \\
| \\
R - Si - O - \\
| \\
R
\end{array}$$

$$\begin{array}{c}
R \\
| \\
Si - O - \\
| \\
R
\end{array}$$

$$\begin{array}{c}
R \\
| \\
R
\end{array}$$

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[wherein R represents a hydrogen atom, hydrocarbon group(s) having 1 to 12 carbon atoms, or  $-OSi(CH_3)_3$  and may be the same or different; and n represents a numeral of 3 or more].

In the general formula (1), the hydrocarbon group(s) having 1 to 12 carbon atoms is/are straight-chain or branched-chain saturated hydrocarbon group(s) and is/are preferably methyl group(s), while n is preferably from 50 to 3000.

Examples of the nonionic silicone polymer represented by the general formula (1)include methyl polysiloxane whose R is entirely methyl group(s).

Examples of the polyether-modified silicone polymer includes those represented by the following general formula (2). Each repeating unit in the silicone polymer of the present invention may be in any form of polymerization such as random polymerization and block polymerization.

$$\begin{array}{c}
CH_{3} \\
R-Si-O- \\
CH_{3} \\
CH_{3}
\end{array}$$

$$\begin{array}{c}
CH_{3} \\
Si-O- \\
CH_{3}
\end{array}$$

$$\begin{array}{c}
CH_{3} \\
CH_{3}
\end{array}$$

$$\begin{array}{c}
CH_{3}$$

$$CH_{3}$$

$$CH$$

[wherein R represents hydrocarbon group(s) having 1 to 12 carbon atoms or group(s) represented by the following general formula (2') and may be the same or different, provided that at least one of R(s) is group(s) represented by the following general formula (2'); m represents 0 or a numeral of 1 or more; and n represents a numeral of 1 or more]

$$\frac{-\left(CH_{2}\right)}{a}\left(OC_{2}H_{4}\right)\left(OC_{3}H_{6}\right)\left(OC_{3}H_{6}\right) = OR \cdot \dots (2')$$

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(wherein R' represents a hydrogen atom or hydrocarbon group(s) having 1 to 10 carbon atoms; a represents a numeral of 1 to 10; b represents a numeral of 1 to 300; and c represents a numeral of 0 to 300)

In the general formula (2), the hydrocarbon group(s) having 1 to 12 carbon atoms represented by R include(s) a straight-chain or branched-chain saturated hydrocarbon group(s). m is preferably from 10 to 500, and n is preferably from 1 to 500. In the general formula (2'), a is preferably 2 to 4, b is preferably from 2 to 50, and c is preferably from 0 to 5.

Examples of the phenyl-modified silicone polymer

includes those represented by the following general formula (3):

$$\begin{array}{c}
CH_{3} \\
R-Si-O- \\
CH_{3} \\
CH_{3}
\end{array}$$

$$\begin{array}{c}
CH_{3} \\
Si-O- \\
CH_{3}
\end{array}$$

$$\begin{array}{c}
CH_{3} \\
Si-O- \\
CH_{3}
\end{array}$$

$$\begin{array}{c}
CH_{3} \\
CH_{3}
\end{array}$$

$$\begin{array}{c}
CH_{3} \\
CH_{3}
\end{array}$$

$$\begin{array}{c}
CH_{3}
\end{array}$$

- [wherein R represents hydrocarbon group(s) having 1 to 12 carbon atoms, -OSi(CH<sub>3</sub>)<sub>3</sub> or phenyl group(s) and may be the same or different, provided that at least one of R(s) is phenyl group(s); m represents 0 or a numeral of 1 or more; and n represents a numeral of 1 or more].
- In the general formula (3), the hydrocarbon group(s) having 1 to 12 carbon atoms represented by R include(s) straight-chain or branched-chain saturated hydrocarbon group(s). m is preferably from 0 to 500, and n is preferably from 1 to 2000.
- As the phenyl-modified silicone polymer represented by the general formula (3) methylphenyl polysiloxane is preferably used.

Examples of the amino-modified silicone polymer includes those represented by the following general formula (4):

20

$$\begin{array}{c}
CH_{3} & CH_{3} \\
| & CH_{3} \\
| & CH_{3}
\end{array}$$

$$\begin{array}{c}
CH_{3} & CH_{3} \\
| & CH_{3}
\end{array}$$

$$\begin{array}{c}
CH_{3} & CH_{3} \\
| & CH_{3}
\end{array}$$

$$\begin{array}{c}
CH_{3} & CH_{3} \\
| & CH_{3}
\end{array}$$

$$\begin{array}{c}
CH_{3} & CH_{3}
\end{array}$$

$$\begin{array}{c}
CH_{3} & CH_{3}
\end{array}$$

$$\begin{array}{c}
CH_{3} & CH_{3}
\end{array}$$

[wherein R represents a hydrogen atom, hydrocarbon group(s) having 1 to 12 carbon atoms, hydroxyl group(s), methoxy group(s) or group(s) represented by the following general formula (4') or (4") and may be the same or different, provided that at least one of R(s) is group(s) represented by the following general formula (4') or (4"); m represents 0 or a numeral of 1 or more; and n represents a numeral of 1 or more

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$$\begin{array}{c} -\left(CH_2\right) + \left(OC_2H_4\right) + \left(OC_3H_6\right) + \left(NHC_2H_4\right) + \left(NH$$

$$\frac{\left(CH_{2}\right)}{a}\left(OC_{2}H_{4}\right)\left(OC_{3}H_{6}\right)\left(NHC_{2}H_{4}\right)N^{+}\left(R'\right)_{3}XZ' \dots (4')$$

(wherein R' represents a hydrogen atom or hydrocarbon group(s) having 1 to 6 carbon atoms and may be the same or different; Z represents a halogen ion or an organic anion; a represents a numeral of 1 to 6; b represents a numeral of 0 to 6; and c represents a numeral of 0 to 6)].

In the general formula (4), the hydrocarbon 20 group(s) having 1 to 12 carbon atoms represented by R

include(s) straight-chain or branched-chain saturated hydrocarbon group(s). m is preferably from 3 to 500, and n is preferably from 1 to 500. In the general formulas (4') and (4"), the hydrocarbon group(s) having 1 to 6 carbon atoms represented by R' include(s) straight-chain or branched-chain saturated hydrocarbon group(s).

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As the amino-modified silicone polymer represented by the said general formula (4), aminodimethycone represented by the following structural formula is preferably used.

$$\begin{array}{c|c} & CH_{3} & & & \\ & & & \\ & & & \\ Si - O & & \\ & & \\ & & CH_{3} & \\ &$$

[wherein m and n are as defined in the general formula (4)]

Examples of the alkyl-modified silicone polymer include, those represented by the following general formula (5):

$$\begin{array}{c} CH_{3} & CH_{3} & CH_{3} \\ | & | & | \\ R-Si-O- & Si-O & Si-O & Si-O & Si-R \\ | & | & | & | \\ CH_{3} & CH_{3} & | & | \\ | & | & | & | \\ CH_{3} & | & | & | \\ \end{array}$$
 ... (5)

20 [wherein R represents hydrocarbon group(s) having 1 to

50 carbon atoms and may be the same or different, provided that at least one of R(s) is hydrocarbon group(s) having 12 to 50 carbon atoms; m represents 0 or a numeral of 1 or more; and n represents a numeral of 1 or more].

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In the general formula (5), the hydrocarbon group(s) having 1 to 50 carbon atoms represented by R include(s) straight-chain or branched-chain saturated hydrocarbon group(s), and hydrocarbon group(s) having 12 to 50 carbon atoms is/are preferably used, and hydrocarbon group(s) having 15 to 40 carbon atoms is/are more preferably used. m is preferably from 10 to 500, and n is preferably from 10 to 500.

Examples of the alkoxy-modified silicone polymer include those represented by the following general formula (6):

$$\begin{array}{c}
CH_{3} \\
| \\
R-Si-O- \\
| \\
CH_{3}
\end{array}$$

$$\begin{array}{c}
CH_{3} \\
| \\
Si-O- \\
| \\
CH_{3}
\end{array}$$

$$\begin{array}{c}
CH_{3} \\
| \\
Si-O- \\
| \\
CH_{3}
\end{array}$$

$$\begin{array}{c}
CH_{3} \\
| \\
CH_{3}
\end{array}$$

[wherein R represents hydrocarbon group(s) having 1 to 20 12 carbon atoms or alkoxy group(s) having 1 to 50 carbon atoms and may be the same or different, provided that at least one of R(s) is alkoxy group(s) having 1 to 50

carbon atoms; m represents 0 or a numeral of 1 or more; and n represents a numeral of 1 or more].

In the general formula (6), the hydrocarbon group(s) having 1 to 12 carbon atoms represented by R include(s) straight-chain or branched-chain saturated hydrocarbon group(s). The alkoxy group(s) having 1 to 50 carbon atoms represented by R include(s) straight-chain or branched-chain saturated alkoxy group(s) and alkoxy group(s) having 1 to 20 carbon atoms is/are preferably used. m is preferably from 3 to 500, and n is preferably from 1 to 100.

Examples of the cyclic silicone polymer includes those represented by the following general formula (7):

10

$$\begin{bmatrix}
CH_3 \\
| \\
Si - O
\end{bmatrix}
\begin{bmatrix}
CH_3 \\
| \\
Si - O
\end{bmatrix}$$
... (7)

[wherein R represents hydrocarbon group(s) having 1 to 12 carbon atoms and may be the same or different in each repeating unit; m represents 0 or a numeral of 1 or more; n represents a numeral of 1 or more; and m + n is from 4 to 10].

In the general formula (7), the hydrocarbon group(s) having 1 to 12 carbon atoms represented by R

include(s) straight-chain or branched-chain saturated hydrocarbon group(s).

The average molecular weight of these silicone polymers is preferably within a range from 100 to 10,000,000, and particularly preferably from 10,000 to 1,000,000.

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The silicone polymer is preferably used in the form of an aqueous solution or a water dispersion in view of the stability for cosmetics.

The incorporation ratio of the amphoteric urethane resin to the silicone polymer is preferably within a range from 0.1/100 to 100/0.01, and particularly preferably from 100/0.01 to 100/5, in terms of a weight ratio.

Furthermore, any components used commonly in cosmetics such as pigments, coloring matters, colorants, perfumes, surfactants, humectants, preservatives, antiseptics, bactericides, antioxidants, oil agents, viscosity modifier and ultraviolet absorbers can be contained in the cosmetic of the present invention, in addition to the amphoteric urethane resin and silicone polymer.

The cosmetics of the present invention can be prepared, for example, by the following methods.

#### Preparation of hair cosmetics (foam hair fixatives)

In the aqueous solution of the amphoteric

urethane resin thus obtained described above, a silicone polymer, various surfactants such as polyoxyethylene alkyl ether, and coconut oil fatty acid diethanolamide, ethanol, deionized water, etc. are blended in the predetermined proportion, and mixed until they are made homogenous to obtain a component X. Then, a component Y made of a propellant (LPG) is added to prepare a desired foam hair fixative.

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# Preparation of hair cosmetics (aerosol spray hair fixative)

In the aqueous solution of the above amphoteric urethane resin, a silicone polymer, deionized water, sodium dioctyl sulfosuccinate, ethanol, etc. are blended in the predetermined proportion, and mixed until they are made homogenous to obtain a component X. Then, a component Y made of a propellant (LPG) is added to prepare a desired aerosol spray hair fixative.

#### Preparation of hair cosmetics (gel-like hair fixatives)

First, a viscosity modifier, triethanolamine, ethanol, deionized water, etc. are blended in the predetermined proportion, and then mixed until a viscous gel is formed to obtain a component X. Then, a silicone polymer, ethanol, deionized water, etc. are blended in the aqueous solution of the amphoteric urethane resin in the predetermined proportion to obtain a component Y. The resulting component Y is added to the above component,

and mixed until they are made homogenous to prepare a desired gel-like hair fixative.

#### Preparation of hair cosmetics (pump spray hair fixatives

In the aqueous solution of the amphoteric urethane resin, a silicone polymer, sodium dioctyl sulfosuccinate, ethanol, deionized water, etc. are blended in the predetermined proportion, and mixed until they are made homogenous to prepare a desired pump spray hair fixative.

10 Furthermore, skin care cosmetics such as shaving cream, skin care lotion and sunscreen lotion; and makeup cosmetics such as nail polish, mascara and foundation can be prepared according to the general preparation methods of these cosmetics.

## 15 Examples

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The following Examples and Comparative Examples further illustrate the present invention in detail.

The following materials were prepared before describing the Examples and Comparative Examples.

#### 20 Amphoteric urethane resin (a)

In a glass four-necked flask equipped with a stirrer, a thermometer, a nitrogen introducing tube and a reflux condenser, 100 g of isophorone disocyanate (IPDI), 60 g of polypropylene glycol (PPG having a molecular weight of 1000), 5 g of cyclohexyl dimethanol

(CHDM), and 38 g of dimethylolbutanoic acid (DMBA) were charged, and then 60 q of ethyl acetate as a solvent was added and the mixture was heated to 80 degree C in an oil bath and allowed to react for four hours. Then, 2 q of N-methyldiethanolamine and 30 g of ethyl acetate were added furthermore, and the mixture was allowed to react for additional three hours. To the resulting mixture, 30 g of polypropylene glycol having an amino group at one end (Jeffamine M1000, manufactured by HUNTSMAN CORPORATION) and 50 g of ethyl acetate were added furthermore, and the mixture was allowed to react for additional one hour to obtain a solution of a polyurethane prepolymer having residual NCO groups. The polyurethane prepolymer having residual NCO groups was dispersed in 750 g of water containing 16 g of potassium hydroxide and then polymerized by the chainextending reaction at 50 degree C for three hours. Ethyl acetate was recovered from the resulting dispersion under reduced pressure to obtain an amphoteric urethane resin which did not substantially contain the solvent.

### Amphoteric urethane resin (b)

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In a glass four-necked flask equipped with a stirrer, a thermometer, a nitrogen introducing tube and a reflux condenser, 100 g of isophorone disocyanate

(IPDI), 60 g of polypropylene glycol (PPG having a molecular weight of 1000), 5 g of cyclohexyl dimethanol (CHDM), 20 g of polyoxyethylene glycol (PEG having a 1000) and, 36 molecular weight of of dimethylolbutanoic acid (DMBA) were charged, and then 60 g of ethyl acetate as a solvent was added, and the mixture was heated to 80 degree C in an oil bath and allowed to react for four hours. Then, 2 g of Nmethyldiethanolamine and 30 g of ethyl acetate were added furthermore, and the mixture was allowed to react for additional three hours. To the resulting mixture, 30 g of polypropylene glycol having an amino group at one end M1000. manufactured by HUNTSMAN (Jeffamine CORPORATION), and 50 q of ethyl acetate were added furthermore, and the mixture was allowed to react for additional one hour to obtain a solution of a polyurethane prepolymer having residual NCO The groups. polyurethane prepolymer having residual NCO groups was dispersed in 750 g of water containing 15 g of potassium hydroxide and then polymerized by the chainextending reaction at 50 degree C for three hours. Ethyl acetate was recovered from the resulting water dispersion under reduced pressure to obtain an aqueous substance of an amphoteric urethane resin which did not

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substantially contain the solvent and had ethylene oxide chain(s) in the structure.

### Amphoteric urethane resin (c)

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In a glass four-necked flask equipped with a stirrer, a thermometer, a nitrogen introducing tube and a reflux condenser, 100 g of isophorone diisocyanate (IPDI), and 3 g of polydimethylsiloxanediol having two OH groups at one end (molecular weight: 1000) were charged, and then the mixture was heated to 80 degree C in an oil bath and allowed to react for two hours. Then, 55 g of polypropylene glycol (PPG having a molecular weight of 1000), 10 g of hydrogenated bisphenol A, and 36 g of dimethylolbutanoic acid (DMBA) were added, and then 60 g of ethyl acetate as a solvent was added, and the mixture was heated to 80 degree C in an oil bath and allowed to react for four hours. Then, 2 g of Nmethyldiethanolamine and 30 g of ethyl acetate were added furthermore, and the mixture was allowed to react for additional three hours. To the resulting mixture, 30 g of polypropylene glycol having an amino group at one end M1000, manufactured by HUNTSMAN (Jeffamine CORPORATION) and 50 g of ethyl acetate were added furthermore, and the mixture was allowed to react for additional one hour to obtain a solution of a polyurethane prepolymer having residual NCO groups. The polyurethane prepolymer having residual NCO groups was dispersed in 750 g of water containing 15 g of potassium hydroxide and then polymerized by the chain-extending reaction at 50 degree C for three hours. Ethyl acetate was recovered from the resulting water dispersion under reduced pressure to obtain an aqueous substance of an amphoteric urethane resin which did not substantially contain the solvent and had dimethylsiloxane chain(s) in the structure.

### 10 Silicone polymer (1) (nonionic)

Methylpolysiloxane (SH200C-2 manufactured by DOW CORNING TORAY SILICONE CO., LTD.)

### Silicone polymer (2) (polyether-modified)

SH3771C manufactured by DOW CORNING

15 TORAY SILICONE CO., LTD.

### Silicone polymer (3) (phenyl-modified)

Methylphenylpolysiloxane (SH556 manufactured by DOW CORNING TORAY SILICONE CO., LTD.)

# Silicone polymer (4) (amino-modified)

20 Amodimethycone (SM8702C manufactured by DOW CORNING TORAY SILICONE CO., LTD.)

### Silicone polymer (5) (alkyl-modified)

KF-412 manufactured by SHIN-ETSU CHEMICAL CO., LTD.

### Silicone polymer (6) (alkoxy-modified)

KF-851 manufactured by SHIN-ETSU CHEMICAL CO., LTD.

### Silicone polymer (7) (cyclic)

5 SH245 manufactured by DOW CORNING TORAY SILICONE CO., LTD.

### Polyoxyethylene stearyl ether

NIKKOL BS-20 manufactured by NIKKO CHEMICALS CO., LTD.

# 10 Coconut oil fatty acid diethanolamide

Amicol CDE-1 manufactured by MIYOSHI OIL & FAT CO., LTD.

### Sodium dioctyl sulfosuccinate

Monawet MO-70E manufactured by MONA 15 INDUSTRIES INC.

### Viscosity modifier

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Alkyl acrylate-polyoxyethylene stearyl ether itaconate copolymer (Structure 2001 manufactured by National Starch and Chemical Company)

# 20 Hair cosmetics (foam hair fixatives)

# Examples 1a to 21a, Comparative Examples 1a to 3a

The respective materials of a component X shown in Tables 1 to 4 described hereinafter were blended in the proportion shown in the same tables and mixed until they were made homogenous to obtain the component X.

Then, a component Y was added in the resulting component X in the proportion shown in the same tables to prepare a foam hair fixative, respectively. The proportion of the amphoteric urethane resin is represented by a dry weight (the same rule applies correspondingly to the following Examples and Comparative Examples).

Using the foam hair fixatives of the Examples and Comparative Examples thus obtained, the respective characteristics were evaluated according to the following criteria. The results are summarized in Tables 1 to 4 described hereinafter.

#### <u>Feel</u>

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0.8 g of the foam hair fixative was applied to a strand of black virgin hairs (having a length of 25 cm and a weight of 5.0 g). Then, the strand of hairs after drying at room temperature was subjected to an organoleptic test using ten panelists and the feel for hair cosmetic was evaluated. Evaluation criteria are set as follows:

: The number of persons, who felt that the strand of hairs is very soft to the feel, is 9 or more

: The number of persons, who felt that the strand of hairs is very soft to the feel, is within a range from 6 to 8.

: The number of persons, who felt that the strand of hairs is very soft to the feel, is within a range from 2 to 5.

x: The number of persons, who felt that the strandof hairs is very soft to the feel, is 1 or less.

### Spread

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0.8 g of the foam hair fixative was applied to a strand of black virgin hairs (having a length of 25 cm and a weight of 5.0 g). Then, the strand was subjected to an organoleptic test using ten panelists and the spread for hair cosmetic was evaluated. Evaluation criteria are set as follows:

: The number of persons, who felt that the spread at the time of application is very good, is 9 or more.

: The number of persons, who felt that the spread at the time of application is very good, is within a range from 6 to 8.

: The number of persons, who felt that the spread at the time of application is very good, is within a range from 2 to 5.

x: The number of persons, who felt that the spread at the time of application is very good, is 1 or less.

#### Touch

0.8 g of the foam hair fixative was applied on a strand of black virgin hairs (having a length of 25 cm and a weight of 5.0 g). Then, the strand of hairs after drying at room temperature was subjected to an organoleptic test using ten panelists and the touch for hair cosmetic was evaluated. Evaluation criteria are set as follows:

: The number of persons, who felt that the strand of hairs after drying is very smooth to the touch, is 9 or more.

: The number of persons, who felt that the strand of hairs after drying is very smooth to the touch, is within a range from 6 to 8.

: The number of persons, who felt that the strand of hairs after drying is very smooth to the touch, is within a range from 2 to 5.

×: The number of persons, who felt that the strand of hairs after drying is very smooth to the touch, is 1 or less.

### 20 Hair washability

0.6 g of the foam hair fixative was applied to black virgin hairs (having a length of 15 cm and a weight of 3 g), followed by drying to make a strand of hairs. After the strand of hairs was slightly loosen using hot water at

40 degree C, 0.4 g of a 10% shampoo solution was applied and the strand was washed for 30 seconds. After the strand was rinsed with hot water at 40 degree C to wash away the shampoo solution, and sufficiently dried at 50 degree C, the hair washability for hair cosmetic were evaluated. Evaluation criteria are set as follows:

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: The number of persons, who felt that the hair washability are very good because the strand after drying has not any setting ability, is 9 or more.

: The number of persons, who felt that the hair washability are very good because the strand after drying has not any setting ability, is within a range from 6 to 8.

: The number of persons, who felt that the hair washability are very good because the strand after drying has not any setting ability, is within a range from 2 to 5.

x: The number of persons, who felt that the hair washability are very good because the strand after drying has not any setting ability, is 1 or less.

(Foam hair Fixative)

		-						
		Examples	SS					
		1a	2a	3a	4a	5a	6a	7a
	Amphoteric urethane resin	3.0	3.0	3.0	3.0	3.0	3.0	3.0
	(Types)	(a)	(a)	(a)	(a)	(a)	(a)	(a)
	Silicone polymer	0.5	0.5	0.5	90	0.5	0.5	0.5
	(Types)	(E)	(2)	(3)	(4)	(2)	(9)	(7)
×	Deionized water	77.2	77.2	77.2	77.2	77.2	77.2	77.2
( Jue	Polyoxyethylene stearyl ether	0.5	0.5	0.5	90	0.5	0.5	0.5
uoc	Ethanol	10.0	10.0	10.0	10.0	10.0	10 0	10.0
Com	Coconut oil fatty acid diethanolamide	0.8	8.0	9.0	0.8	0.8	9.0	8 0
ပိ	Component Y Propellant (LPG)	8.0	8.0	8.0	8.0	8.0	8 0	8.0
Feel	el							
Sp	Spread							
ို	Touch							
Ha	Hair washability							

Table 2

(Foam Hair Fixative)

			Examples	3S					
			8a	9a	10a	11a	12a	13a	14a
	Amphoteric urethane resin	ethane resin	3.0	3.0	3.0	3.0	3.0	3.0	3.0
	(Types)		(Q)	. (q)	(q)	(q)	(q)	(q)	(q)
	Silicone polymer	er	0.5	0.5	9.0	9.0	9.0	0.5	0.5
	(Types)		Ξ.	(2)	(3)	(4)	(2)	(9)	(2)
X	Delonized water	31	77.2	77.2	77.2	77.2	77.2	77.2	77.2
( lue	Polyoxyethylene stearyl ether	le stearyl ether	0.5	0.5	0.5	0.5	0.5	0.5	90
uoc	Ethanol		10.0	10 0	10.0	10.0	10 0	10.0	10 0
Com	Coconut oil fatty acid diethanolamide	ly acid	0.8	8.0	8.0	9.0	9.0	9.0	9.0
Š	Component Y	Propellant (LPG)	8 0	8.0	8.0	8.0	8.0	8.0	8.0
Feel	-								
Spr	Spread								
Touch	hor								
На	Hair washability								

Table 3

(Foam Hair Fixative)

			Examples	es					
		•	15a	16a	17a	18a	19a	20a	21a
	Amphoteric urethane resin	ethane resin	3.0	3.0	3.0	3.0	3.0	3.0	3.0
	(Types)	:		(c)	(c)	(c)	(c)	(c)	(c)
	Silicone polymer	er	9.0	0.5	9.0	6.0	6.0	0.5	0.5
	(Types)	:	Ξ	(2)	(3)	(4)	(2)	(9)	(2)
X	Deionized water	je	77.2	77.2	2.77	77.2	27.7	77.2	77.2
tna	Polyoxyethylene stearyl ether	ie stearyl ether	9 0	9.0	9.0	9.0	0.5	0.5	9.0
uod	Ethanol		10.0	10.0	10.0	10.0	10.0	10.0	10.0
Com	Coconut oil fatty acid diethanolamide	ly acid	0.8	9.0	0.8	8.0	0.8	9.0	9.0
Š	Component Y	Propellant (LPG)	8.0	8.0	8.0	8.0	8.0	8.0	8.0
Fee	-								
Spr	Spread								
Touch	yor								
Ha	Hair washability								

(Foam Hair Fixative)

(Product)

			Compan	Comparative Examples	nples
			1a	2a	3а
	Amphoteric urethane resin	nane resin	3.0	3.0	3.0
	(Types)	:	(a)	(q)	(c)
	Silicone polymer		ı	1	1
	(Types)		1		1
×	Delonized water		77.77	77.7	77.7
( Jue	Polyoxyethylene stearyl ether	stearyl ether	0.5	0.5	0.5
uoc	Ethanol		10.0	10.0	10.0
Comp	Coconut oil fatty acid diethanolamide	acıd	0.8	0.8	8.0
Õ	Component Y	Propellant (LPG)	8.0	8.0	8.0
Feel	<u> </u>				
Spi	Spread				
Tol	Touch				
Ha	Haır washability				

As is apparent from the results shown in Tables 1 to 4, the foam hair fixatives of the Examples have very good feel and good spread at the time of application and are superior in touch and hair washability because the amphoteric urethane resin and silicone polymer are used in combination. It is also apparent that the foam hair fixatives of the Examples using the amphoteric urethane resin (b) having ethylene oxide chain(s) in its structure are markedly superior in hair washability. It is also apparent that the foam hair fixatives of the Examples using the amphoteric urethane resin (c) having polysiloxane bonds in its structure are markedly superior in touch.

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To the contrary, the foam hair fixatives of the Comparative Examples have good feel because the amphoteric urethane resin is used, but are inferior in spread at the time of application.

### Hair cosmetics (aerosol spray hair fixatives)

### Examples 1b to 21b, Comparative Examples 1b to 3b

The respective materials of a component X shown in Tables 5 to 8 described hereinafter were blended in the proportion shown in the same tables and mixed until they are made homogenous to obtain the component X. Then, a component Y was added in the resulting

component X in the proportion shown in the same tables to prepare an aerosol spray hair fixative, respectively.

Using the aerosol spray hair fixatives of the Examples and Comparative Examples thus obtained, the respective characteristics were evaluated according to the criteria for hair cosmetics. The results are summarized in Tables 5 to 8 described hereinafter.

(Aerosol spray hair fixative)

			Examples	es					
			1p	2b	36	4b	2p	<b>q</b> 9	7b
	Amphoteric urethane resin		3.0	3.0	3.0	3.0	3.0	3.0	3.0
	(Types)		(a)	(a)	(a)	(a)	(a)	(a)	[(a)
	Silicone polymer	ıer	0.5	0.5	0.5		0.5	0.5	0.5
X 1n	(Types)		(1)	(2)	(3)	(4)	(2)	(9)	(7)
əuo	Deionized waier	er	0.7	7.0	7.0	7.0	7.0	7.0	7.0
dwo		Sodium dioctyl sulfosuccinate	0.3	0.3	0.3	0.3	0.3	0.3	0.3
ာ၁	Ethanol		49.2	49.2	49.2	49.2	49.2	49.2	49.2
ଓ	Component Y	Propellant (LPG)	40.0	40.0	40 0	40.0	40.0	40.0	40.0
Feel	<u>a</u>								
Spi	Spread								
Toı	Touch								
Hai	Hair washability								

(Aerosol spray hair fixative)

Table 6

			Examples	es					
			8b	q6	10b	116	12b	13b	14b
	Amphoteric	Amphoteric urethane resin	3.0	3.0	3.0	3.0	3.0	3.0	3.0
	(Types)	:	: (a)	(q)	: (a)	(q)	(a)	(q)	( <u>a</u> )
	Silicone polymer	lymer	0.5	0.5	0.5	0.5	0.5	0.5	0.5
X 1n	(Types)		(1)	(2)	(3)	(4)	(2)	(6)	. (2)
əuo	Deionized water	water	7.0	7.0	7.0	7.0	7.0	7.0	7.0
dш	Sodium dio	Sodium dioctyl sulfosuccinate	0.3	0.3	0.3	0.3	0.3	0.3	0.3
ာ	Ethanol		49.2	49.2	49.2	49.2	49.2	49.2	49.2
Comp	Component Y	Propellant (LPG)	40.0	40 0	40.0	40.0	40.0	40.0	40.0
Feel									
Spread	þ								
Touch									
Hairw	Haır washability								

Table 7

(Aerosol spray hair fixative)

			Examples	es					
			15b	16b	17b	18b	19b	20b	21b
	Amphoteric urethane resin	sthane resin	3.0	3.0	3.0	3.0	3.0	3.0	3.0
	(Types)		(0)	(0)	(c)	(c)	(c)	(c)	(o)
	Silicone polymer	er	0.5	0.5	0.5	9.0	0.5	0.5	9 0
X Ju	(Types)	-	(1)	(2)	(3)	(4)	(5)	(9)	(7)
əuo	Deionized water	91	7.0	7.0	0.2	0.7	7.0	7.0	7.0
dwo	Sodium dioctyl	Sodium dioctyl sulfosuccinate	0.3	0.3	0.3	0.3	0.3	0.3	0.3
ာ	Ethanol		49.2	49.2	49.2	49.2	49.2	49.2	49.2
Co	Component Y	Propellant (LPG)	40.0	40.0	40.0	40.0	40.0	40.0	40 0
Feel	-								
Spr	Spread								
Touch	ıch								
Hai	Hair washability								

Table 8

(Aerosol spray hair fixative)

		3	mpara	Comparative Examples	mples
		10		2b	3p
	Amphoteric urethane resin	3.0		3.0	3.0
	(Types)	(a)		(b)	(c)
	Silicone polymer	1		1	-
X in	(Types)			.	
əuo	Deionized water	7.0	0	7.0	0.7
dwo	Sodium dioctyl sulfosuccinate	0.3	3	0.3	0.3
ာ	Éthanol	49.7	7.	49.7	49.7
Co	Component Y Propellant (LPG)	40.0	0.	40 0	40.0
Feel	-				
Spr	Spread				
Toı	Touch				
Наі	Hair washability				

As is apparent from the results shown in Tables 5 to 8, the aerosol spray hair fixatives of the Examples have very good feel and good spread at the time of application and are superior in touch and hair washability because the amphoteric urethane resin and silicone polymer are used in combination. It is also apparent that the aerosol spray hair fixatives of the Examples using the amphoteric urethane resin (b) having ethylene oxide chain(s) in its structure are markedly superior in hair washability. It is also apparent that the aerosol spray hair fixatives of the Examples using the amphoteric urethane resin (c) having polysiloxane bond(s) in its structure are markedly superior in touch.

To the contrary, the aerosol spray hair fixatives of the Comparative Examples have good feel because the amphoteric urethane resin is used, but are inferior in spread at the time of application.

### Hair cosmetics (jerry hair fixatives)

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### Examples 1c to 21c, Comparative Examples 1c to 3c

The respective materials of a component X shown in Tables 9 to 12 described hereinafter were blended in the proportion shown in the same tables and mixed until viscous gel is formed to obtain the component X. Then, a component Y which was prepared by blending the respective materials in the proportion shown in the same

table was added in the resulting component X and mixed until they are made homogenous to prepare a jelly hair fixative, respectively.

Using the jerry hair fixatives of the Examples and

Comparative Examples thus obtained, the respective characteristics were evaluated according to the criteria for hair cosmetics. The results are summarized in Tables 9 to 12 described hereinafter.

(Gel-tike hair fixative)

		Examples	ss					
		10	2c	3с	4c	<b>5</b> c	90	7c
	Thickener	1.5	1.5	1.5	1.5	1.5	1.5	15
X	Triethanolamine	1.1	1.1	1.1	1.1	1.1	1.1	11
poneni	Ethanol	5.0	5.0	5.0	5.0	5.0	5.0	5.0
Com	Deionized water	90.09	90.09	90.09	50.0	50.0	90.09	50.0
	Amphoteric urethane resin	3.0	3.0	3.0	3.0	3.0	3.0	3.0
	(Types)	(a)	(a)	(a)	(a)		(a)	(a)
ΥJu	Silicone polymer	0.5	0.5	0.5	9.0	9.0	0.5	0.5
iəuo	(Types)	E	(2)	(3)	(4)	(5)	(6)	(7)
du	Ethanol	5.0	5.0	5.0	5.0	5.0	5.0	2.0
ာ၁	Deionized water	33.9	33.9	33 9	33.9	33 9	33.9	33.9
Feel								
Spr	Spread							
Touch	ıch							
Ī	Hair washability							

Table 10

(Gel-like hair fixative)

		Examples	se					
		вс	96	10c	110	12c	13c	14c
	Thickener	1.5	1.5	1.5	1.5	15	1.5	1.5
X	Triethanolamine	1:	1.1	1.1	1.1	1.1	1.1	1.1
boueu	Ethanol	5.0	2 0	5.0	5.0	5.0	9.0	5.0
Com	Deionized water	50.0	50.0	0.03	20.0	90.09	90.09	20.0
	Amphoteric urethane resin	3.0	3.0	3.0	3.0	3.0	3.0	3.0
	(Types)	(Q)	(a)	(a)	(q)	. (q)	(q)	(q)
Υ'n	Silicone polymer	0.5	0.5	0.5	0.5	9.0	9.0	0.5
əuo	(Types)	; : E	(2)	(3)	(4)	(2)	(9)	(2)
dш	Ethanol	5.0	5.0	5.0	5.0	5.0	5.0	5.0
ာ	Deionized water	33.9	33 9	33.9	33.9	33.9	33 9	33.9
Feel	- To							
Spr	Spread							
Touch	yor							
Hai	Hair washability							

Table 11

(Gel-like hair fixative)

		Examples	se					
		15c	16c	17c	18c	19c	20c	21c
	Thickener	1.5	1.5	1.5	1.5	1.5	15	1.5
X	Triethanolamine	1.1	1.1	1.1	1.1	1.1	1.1	1.1
boueu	Ethanol	5.0	5.0	5.0	5.0	5.0	5.0	5.0
Com	Deionized water	50 0	50.0	50.0	50.0	20 0	20.0	90.09
	Amphoteric urethane resin	3.0	3.0	3.0		3.0	3.0	3.0
	(Types)	(C)	(c)		(c)	(c)	(c)	(c)
Υıυ	Silicone polymer	0.5	0.5	0.5	0.5	0.5	0.5	0.5
əuo	(Types)	E	(2)	(3)	(4)	(2)	(9)	(2)
dwo	Ethanol	5.0	5.0	5.0	5.0	5.0	20	9.0
ာ၁	Deionized water	33.9	33.9	33 9	33.9	33.9	33.9	33 9
Feel								
Spr	Spread							
Touch	ich							
Hai	Hair washability							

Table 12

(Gel-like hair fixative)

		Compar	Comparative Examples	nples
		1c	2c	3c
	Thickener	1.5	15	1.5
ΧI	Triethanolamine	1.1	1.1	1.1
uəuod	Ethanol	5.0	5.0	5.0
Com	Deionized water	50.0	50.0	20 0
	Amphoteric urethane resin	3.0	3.0	3.0
	(Types)	(a)	(q)	(c)
Y In	Silicone polymer	:	1	1
əuc	(Types)	: :		
dwo	Ethanol	5.0	5.0	9.0
ာ၁	Deionized water	34.4	34.4	34.4
Feel				
Spr	Spread			
Touch	ich			
Hai	Hair washability			

As is apparent from the results shown in Tables 9 to 12, the jelly hair fixatives of the Examples have very good feel and good spread at the time of application and are superior in touch and hair washability because the amphoteric urethane resin and silicone polymer are used in combination. It is also apparent that the gel-like hair fixatives of the Examples using the amphoteric urethane resin (b) having ethylene oxide chain(s) in its structure are markedly superior in hair washability. It is also apparent that the gel-like hair fixatives of the Examples using the amphoteric urethane resin (c) having polysiloxane bond(s) in its structure are markedly superior in touch.

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To the contrary, the gel-like hair fixatives of the Comparative Examples have good feel because the amphoteric urethane resin is used, but are inferior in spread at the time of application.

### Hair cosmetics (pump spray hair fixatives)

# Examples 1d to 21d, Comparative Examples 1d to 3d

The respective materials shown in Tables 13 to 16 described hereinafter were blended in the proportion shown in the same table and mixed until they are made homogenous to prepare a pump spray hair fixative, respectively.

Using the pump spray hair fixatives of the Examples and Comparative Examples thus obtained, the respective characteristics were evaluated according to the criteria for hair cosmetics. The results are summarized in Tables 13 to 16 described hereinafter.

(Pump spray hair fixative)

1d     2d     3d     4d       hane resin     3.0     3.0     3.0     3.0       r     (a)     (a)     (a)     (a)     3.0       r     0.5     0.5     0.5     0.5       (1)     (2)     (3)     (4)       sulfosuccinate     0.3     0.3     0.3     0.3       100     100     10.0     10.0       86.2     86.2     86.2     86.2		Examples	es					
boteric urethane resin 3.0 3.0 3.0 3.0 es) es) es) one polymer 0.5 0.5 0.5 0.5 es) es) um dioctyl sulfosuccinate 0.3 0.3 0.3 0.3 ond not on the color of the colo		10	2d	39	4d	5d	p9	74
es)  one polymer  one polymer  es)  in dioctyl sulfosuccinate  ond  onized water  ad  one polymer  one polymer  one polymer  one polymer  one polymer  one one polymer  one one polymer  one one polymer  one	Amphoteric urethane resin	3.0	3.0	3.0	3.0	3.0	3.0	3.0
bes) um dioctyl sulfosuccinate 0.5 0.5 0.5 0.5 (1) (2) (3) (4) (1) (2) (3) (4) (4) (2) (3) (4) (5) (6) (6) (6) (7) (10) (10) (10) (8) (2) (8) (2) (8) (2) (9) (10)	(Types)	(a)	(a)	(a)	(a)	(a)	(a)	(a)
es) (1) (2) (3) (4) (4) um dioctyl sulfosuccinate 0.3 0.3 0.3 0.3 0.3 0.3 0.3 nol nol 10.0 10.0 10.0 10.0 nized water 86.2 86.2 86.2 86.2 ad	Silicone polymer	0.5	0.5	0.5	0.5	0.5	0.5	0.5
um dioctyl sulfosuccinate         0.3         0.3         0.3         0.3           nol         10.0         10.0         10.0         10.0           nized water         86.2         86.2         86.2         86.2           ad         h         h         h         h	(Types)	ΞΞ	(2)	(3)	(4)	(2)	(9)	(2)
noi 10.0 10.0 10.0 10.0 nized water 86.2 86.2 86.2 86.2 86.1 86.2 86.2 86.2 86.2 86.2 8d.2 8d.2 8d.2 8d.2 8d.2 8d.2 8d.2 8d	Sodium dioctyl sulfosuccinate	0.3	0.3	0.3	03	0.3	0.3	0.3
nized water 86.2 86.2 86.2 86.2 ad ad the second se	Ethanoi	100	10.0	10.0	10.0	10.0	10.0	10 0
Spread Touch Har washability	Deionized water	86.2	86.2	86.2	86.2	86.2	86.2	86.2
Spread Touch Hair washability	Feel							
Touch Hair washahility	Spread							
Hair washahility	Touch							
	Hair washability							

(Pump spray hair fixative)

	Examples	Se					
	84	p6	10d	11d	12d	13d	14d
Amphoteric urethane resin	3.0	30	3.0	3.0	3.0	3.0	3.0
(Types)	( <u>a</u>	<b>(a)</b>	[q)	(q)	(q)	(q)	(q)
Silicone polymer	9.0	0.5	0.5	0.5	0.5	0.5	0.5
(Types)	(1)	[(2)	(3)	(4)		(9)	(2)
Sodium dioctyl sulfosuccinate	0.3	0.3	0.3	0.3	0.3	0.3	0.3
Ethanol	100	10.0	10.0	10.0	10.0	10 0	10.0
Deionized water	86 2	86.2	86.2	86.2	86.2	86.2	86 2
Feel							
Spread							
Touch							
Hair washability							

(Pump spray hair fixative)

	Examples	se					
	15d	16d	17d	18d	19d	20d	21d
Amphoteric urethane resin	3.0	3.0	3.0	3.0	3.0	3.0	3.0
(Types)	(c)	: : (0)	(c)	(0)	(c)	(c)	(c)
Silicone polymer	0.5	0.5	0.5	0.5	90	9.0	9.0
(Types)	· (E)	. (2)	(3)	(4)	(2)	(9)	(7)
Sodium dioctyl sulfosuccinate	0.3	0.3	0.3	0.3	0.3	0.3	0.3
Ethanol	10.0	10.0	10.0	10.0	10.0	10.0	10.0
Deionized water	86.2	86.2	86.2	86.2	86.2	86.2	86 2
Feel							
Spread							
Touch							
Hair washability							

Table 16

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	Compar	Comparative Examples	mples
	1d	2d	ρε
	3.0	3.0	3.0
(Types)	(a)	(q)	(c)
Silicone polymer		-	-
(Types)		. 1	
Sodium dioctyl sulfosuccinate 0	0.3	0.3	0.3
Ethanol	10.0	10.0	10.0
Deionized water	86.7	86.7	86.7
Feel			
Spread			
Touch			
Hair washability			

As is apparent from the results shown in Tables 13 to 16, the pump spray hair fixatives of the Examples have very good feel and good spread at the time of application and are superior in touch and hair washability because the amphoteric urethane resin and silicone polymer are used in combination. It is also apparent that the pump spray hair fixatives of the Examples using the amphoteric urethane resin (b) having ethylene oxide chain(s) in its structure are markedly superior in hair washability. It is also apparent that the pump spray hair fixatives of the Examples using the amphoteric urethane resin (c) having polysiloxane bond(s) in its structure are markedly superior in feel.

To the contrary, the pump spray hair fixatives of the Comparative Examples have good feel because the amphoteric urethane resin is used, but are inferior in spread at the time of application.

# Skin care cosmetics (skin care lotions)

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### Examples 1e to 7e, Comparative Example 1e

The respective materials of a component X shown in Tables 17 and 18 described hereinafter were blended in the proportion shown in the same tables and heated to 80 degree C to obtain the component X. The respective materials of a component Y were blended in the proportion shown in the same tables and heated to 80

degree C to obtain the component Y. Then, the component X and component Y were mixed, followed by stirring at 80 degree C for 30 minutes. A viscosity modifier was added, and mixed until they were made homogenous, then cooled down to 40 degree C to prepare a skin care lotion, respectively.

Using the skin care lotions of the Examples and Comparative Example thus obtained, the respective characteristics were evaluated according to the following criteria. The results are summarized in Tables 17 and 18 described hereinafter.

#### Feel

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The feel for skin care cosmetic was evaluated at a practical use test using ten panelists. Evaluation criteria are set as follows:

: The number of persons, who did not feel tight and also felt soft, is 9 or more.

: The number of persons, who did not feel tight and also felt soft, is within a range from 6 to 8

: The number of persons, who did not feel tight and also felt soft, is within a range from 2 to 5.

x: The number of persons, who did not feel tight and also felt soft, is 1 or less

### Spread

The spread for skin care cosmetic was evaluated by a practical use test using ten panelists. Evaluation criteria are set as follows:

- 5 : The number of persons, who felt that the spread at the time of application is very good, is 9 or more.
- : The number of persons, who felt that the spread at the time of application is very good, is within a 10 range from 6 to 8.
  - : The number of persons, who felt that the spread at the time of application is very good, is within a range from 2 to 5.
- x: The number of persons, who felt that the 15 spread at the time of application is very good, is 1 or less.

(Skin care lotion)

		Examples	es		
		1e	2e	Зе	4e
	Octyl methoxycinnamate	7.5	7.5	7.5	7.5
	Ether polyoxystearate	10	1.0	1.0	1.0
	Emulsion type glyceryl stearate	1.0	1.0	1.0	1.0
X	Stearic acid	1.5	1.5	1.5	1.5
poneni	Mixture of titanium dioxide and C <sub>12-15</sub> alkyl benzoate	1.7	1.7	1.7	1.7
Com	Polyoxyethylene-added dimethycone	0.5	0.5	0.5	0.5
	Amphoteric urethane resin (a)	1.0	1.0	10	1.0
		0.3	0.3	0.3	0.3
	(Types)	(1)	(2)	(3)	(4)
Y in	Deionized water	61.5	61.5	615	615
əuo	Triethanolamıne (99%)	4.0	4.0	4.0	4 0
dwo	Antiseptic	q.s.	q.s.	q.s.	d s.
ာ၁	Thickener (2%)	20 0	20.0	20.0	20 0
Feel					
Spr	Spread				

(Skin care lotion)

		Examples			Comparative Example
		5e	ee	7е	1e
	Octyl methoxycinnamate	7.5	2.5	7.5	7.5
	Ether polyoxystearate	1.0	1.0	1.0	1.0
	Emulsion type glyceryl stearate	1.0	1.0	1.0	1.0
X	Stearic acid	1.5	1.5	1.5	1.5
uəuod	Mixture of titanium dioxide and C <sub>12-15</sub> alkyl benzoate	1.7	1.7	1.7	1.7
Com	Polyoxyethylene-added dimethycone	0.5	9:0	9:0	0.5
	Amphoteric urethane resin (a)	1.0	1.0	1.0	1.0
	Silicone polymer	0.3	0.3	0.3	
	(Types)	(2)	(9)	(2)	
Υ'n	Deionized water	61.5	61.5	61.5	61.8
əuo	Triethanolamine (99%)	4.0	4.0	4.0	4.0
dwo	Antiseptic	q s.	q.s.	q.s.	q s.
ာ၁	Thickener (2%)	20 0	20.0	20.0	20.0
Feel	-				
Spr	Spread				,

As is apparent from the results shown in Tables 17 and 18, the skin care lotions of the Examples have very good feel and good spread at the time of application because the amphoteric urethane resin and silicone polymer are used in combination.

To the contrary, the skin care lotion of the Comparative Example has good feel because the amphoteric urethane resin is used, but is inferior in spread at the time of application.

## 10 Skin care cosmetics (shaving creams)

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# Examples 1f to 7f, Comparative Example 1f

The respective materials of a component X shown in Tables 19 and 20 described hereinafter were blended in the proportion shown in the same tables and heated to 80 degree C to obtain the component X. The respective materials of a component Y were blended in the proportion shown in the same tables and heated to 80 degree C to obtain the component Y. Then, the component X and component Y were mixed at 80 degree C and cooled down to 40 degree C. An antiseptic and a perfume were added in a sufficient quantity to prepare shaving cream, respectively.

Using the shaving creams of the Examples and Comparative Example thus obtained, the respective characteristics were evaluated according to the criteria

for skin care cosmetics. The results are summarized in Tables 19 and 20 described hereinafter.

y.

(Shaving cream)

		Examples	Se		
		1f	2f	3f	4f
	Stearic acıd	8.0	8.0	8.0	8.0
X	Mineral oil	2.0	2.0	2.0	2.0
uəuod	Isopropyl myristate	2.0	2.0	20	2.0
Соті	Glyceryl stearate	6.0	6.6	0.5	0.5
	Amphoteric urethane resin (a)	0.5	0.5	0.5	0.5
		0.3	0.3	0.3	0.3
Υ'n	(Types)	Ξ	(2)	(3)	· (4)
əuc	Deionized water	72.5	72.5	72.5	72.5
dш	Thickeners (2%)	10.0	10.0	10.0	10.0
၁	Triethanolamine (99%)	4.2	4.2	4.2	4.2
Feel	-				
Spr	Spread				

(Shaving cream)

					The second secon
		Examples	S		Comparative Example
		5f	6f	7.f	1f
	Stearic acıd	8 0	8.0	8.0	8.0
X	Mineral oil	2.0	2.0	2.0	2.0
uəuod	Isopropyl myristete	2.0	2.0	2.0	2.0
mo)	Glyceryl stearate	9.5	0.5	9.5	0.5
	Amphoteric urethane resin (a)	0.5	0.5	0.5	0.5
	Silicone polymer	0.3	0.3	03	
J 1U	(Types)	(2)	(9)	(7)	
əuc	Deionized water	72.5	72.5	72.5	72.8
dw	Thickeners (2%)	10.0	10.0	10.0	10.0
S	Triethanolamine (99%)	4.2	4.2	4.2	4.2
Feel	10				
Spr	Spread				

As is apparent from the results shown in Tables 19 and 20, the shaving creams of the Examples have very good feel and good spread at the time of application because the amphoteric urethane resin and silicone polymer are used in combination.

To the contrary, the shaving cream of the Comparative Example has good feel because the amphoteric urethane resin is used, but is inferior in spread at the time of application.

#### 10 Skin care cosmetics (sunscreen lotions)

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# Examples 1q to 7q, Comparative Example 1q

The respective materials of a component X shown in Tables 21 and 22 described hereinafter were blended in the proportion shown in the same tables and heated to 80 degree C to obtain the component X. The respective materials of a component Y were blended in the proportion shown in the same tables and heated to 80 degree C to obtain the component Y. Then, the component X and component Y were mixed at 80 degree C to prepare a sunscreen lotion, respectively.

Using the sunscreen lotions of the Examples and Comparative Example thus obtained, the respective characteristics were evaluated according to the criteria for skin care cosmetics. The results are summarized in Tables 21 and 22 described hereinafter.

(Sunscreen lotion)

		Examples	SS		
		19	29	39	49
	Octyl cinnamate	7.50	7.50	7.50	7.50
	Octyl palmıtate	5.00	5.00	5.00	5.00
,×	Cetyl alcohol	1.00	1.00	1.00	1.00
tna	Polyethylene glycol monostearate	1.50	1.50	1.50	1.50
uodwo	Poly(oxyethylene oxypropylene) methyl-polysiloxane copolymer	1.00	1.00	1.00	1.00
ာ၁	Dimethylstearylamine	2.00	2.00	2.00	2.00
	Amphoteric urethane resin (a)	1.00	1.00	1.00	1.00
	Silicone polymer	0.50	0.50	0.50	0.50
		Ξ	(2)	(3)	(4)
	Purified water	69.5	69.5	5.69	69.5
		ည	5	2	2
Y Ju	Triethanolamine (99%)	0.70	0.70	0.70	0.70
oue	Thickeners (2%)	10.0	10.0	10.0	10.0
dwo		0	0	0	0
ာ၁	Antiseptic	0.25	0.25	0.25	0 25
Fee					
Spr	Spread				

(Sunscreen lotion)

		Examples	Š		Comparative Example
		59	6g	7g	1g
	Octyl cınnamate	7.50	7.50	7.50	7.50
	Octyl palmitate	5.00	5.00	5 00	5 00
X	Cetyl alcohol	1 00	1.00	1.00	1.00
( Jua	Polyethylene glycol monostearate	1 50	1.50	1.50	1.50
uoduo	Poly(oxyethylene oxypropylene) methyl-polysiloxane copolymer	1.00	1.00	1.00	1.00
ာ	Dimethylstearylamine	2.00	2.00	2.00	2.00
	Amphoteric urethane resin (a)	1 00	1.00	1.00	1.00
	Silicone polymer	0.50	0.50	09.0	1
	(Types)	(5)	(9)	. (2)	
	Purified water	69.5	69.5	69.5	70.0
		5	5	5	9
Υtη	Triethanolamine (99%)	0.70	0.70	0.70	0.70
əuc	Thickeners (2%)	10 0	10.0	10.0	10.0
dwo		0	0	0	0
၁၁	Antiseptic	0.25	0 25	0 25	0 25
Feel	1				
Spr	Spread				
	A 44 - 14 - 14 - 14 - 14 - 15 - 16 - 16 - 16 - 16 - 16 - 16 - 16				

As is apparent from the results shown in Tables 21 and 22, the sunscreen lotions of the Examples have very good feel and good spread at the time of application because the amphoteric urethane resin and silicone polymer are used in combination.

To the contrary, the sunscreen lotion of the Comparative Example has good feel because the amphoteric urethane resin is used, but is inferior in spread at the time of application.

# 10 Make-up cosmetics (nail polishs)

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# Examples 1h to 7h, Comparative Example 1h

As shown in Tables 23 and 24 described hereinafter, a pigment was dispersed in deionized water in the proportion shown in the same tables and other components were added in the proportion shown in the same tables. The resultant was mixed until it was made homogenous with stirring and deaerated to prepare a nail polish, respectively.

Using the nail polishs of the Examples and 20 Comparative Example thus obtained, the respective characteristics were evaluated according to the following criteria. The results are summarized in Tables 23 and 24 described hereinafter.

#### <u>Feel</u>

The feel for skin care cosmetic was evaluated at a practical use test using ten panelists. Evaluation criteria are set as follows:

5 : The number of persons, who did not feel tight and also felt soft, is 9 or more.

: The number of persons, who did not feel tight and also felt soft, is within a range from 6 to 8.

: The number of persons, who did not feel tight and also felt soft, is within a range from 2 to 5.

x: The number of persons, who did not feel tight and also felt soft, is 1 or less.

#### Spread

The spread for skin care cosmetic was evaluated

15 at a practical use test using ten panelists. Evaluation

criteria are set as follows:

: The number of persons, who felt that the spread at the time of application is very good, is 9 or more.

20 : The number of persons, who felt that the spread at the time of application is very good, is within a range from 6 to 8.

- : The number of persons, who felt that the spread at the time of application is very good, is within a range from 2 to 5.
- x: The number of persons, who felt that the spread at the time of application is very good, is 1 or less.

Table 23

(Naıl Polish)

		Examples	SS		
		1h	2h	3h	4h
	Amphoteric urethane resin (a)	10.0	10.0	10.0	10.0
əse			0.5	0.5	0.5
syd	(Types)	(1)	(2)	(3)	(4)
ater	Delonized water	86.1	86.1	86.1	86.1
M	Bentonite	9.0	9.0	9.0	9.0
Pig	Pigment	2.5	2.5	2.5	2.5
	Perfume	0.1	0.1	0.1	0.1
hera	Antiseptic	0.1	0.1	0.1	0.1
10	Silicone defoamer	0.1	0.1	0.1	0.1
Feel					
Spr	Spread				

(Naıl polish)

		Examples	s		Comparative Example
		5h	9	7h	1h
	Amphoteric urethane resin (a)	10.0	10.0	10.0	10.0
əsı	Silicone polymer	0.5	0.5	0.5	ı
eud	(Types)	(2)	(9)	(2)	1
ater	Deionized water	86.1	86.1	86.1	86.1
M	Bentonite	9.0	9.0	9.0	9.0
Pig	Pigment	2.5	2.5	2.5	2.5
5	Perfume	0.1	0.1	0.1	0.1
hers	Antiseptic	0.1	0.1	0.1	0.1
10	Silicone defoamer	0.1	0.1	0.1	0.1
Feel	4				
Spr	Spread				

As is apparent from the results shown in Tables 23 to 24, the nail polishes of the Examples have very good feel and good spread at the time of application because the amphoteric urethane resin and silicone polymer are used in combination.

To the contrary, the nail polish of the Comparative Example has good feel because the amphoteric urethane resin is used, but is inferior in spread at the time of application.

#### 10 Make-up cosmetics (mascaras)

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#### Examples 1i to 7i, Comparative Example 1i

As shown in Tables 25 and 26 described hereinafter, propylene glycol, triethanolamine, a viscosity modifier, and an antiseptic were blended with purified water in the proportion shown in the same tables and, after dissolving them at 80 degree C, the pigment was dispersed therein to form a water phase. Then, the respective materials of a component Z were blended in the proportion shown in the same tables and dissolved at 80 degree C to form an oil phase. The oil phase was aded to the water phase and the mixture was emulsified by using a homogenizing mixer. Then, the respective materials of a component Y were gradually charged in the proportion shown in the same tables, followed by stirring

using a homogenizing mixer and further cooled down to room temperature to prepare a mascara, respectively.

Using the mascaras of the Examples and Comparative Example thus obtained, the respective characteristics were evaluated according to the criteria for make-up cosmetics. The results are summarized in Tables 25 and 26 described hereinafter.

(Mascara)

			Examples	Si		
			11	21	3i	4i
	Propylene glycol	glycol	20	5.0	5.0	5.0
	Trethanolamine	amine	1.0	1.0	1.0	10
	Thickener (2%)	(2%)	10.0	10.0	10.0	10 0
X	Antiseptic		0.5	0.5	0.5	0.5
tnənoo	Pigment	and the state of t	10 0	10.0	10.0	10.0
Comp	Purified water	ater	51.0	510	51.0	510
	Amphoter	Amphoteric urethane resin (a)	10 0	10 0	100	10 0
Y Jna	Silicone polymer	olymer	0.5	0.5	9:0	0.5
Compon	(Types)		(E)	. (2)	(3)	(4)
S	nponent	Stearic acid	0.9	09	0.9	0.9
7	. 7	Beeswax	0.9	0.9	0.9	0.9
Feel	_					
Spr	Spread					

(Mascara)

L			Examples	s		Comparative Example
			51	ej	7i	- II
	Propylene glycol	e glycol	5.0	9.0	5.0	5.0
	Triethanolamine	lamine	1.0	1.0	1.0	1.0
	Thickener (2%)	r (2%)	10.0	10.0	10.0	10.0
X	Antiseptic		6.5	0.5	0.5	0.5
boueu	Pigment		10.0	10.0	10.0	10.0
moD	Purified water	/ater	51.0	51.0	51.0	51.0
	Amphote	Amphoteric urethane resin (a)	10.0	10.0	10.0	10.0
Y Inen	Silicone polymer	oolymer	0.5	0.5	0.5	ı
Compo	(Types)		(5)	(e) 	(7)	
So	Component	Stearic acid	0.9	6.0	6.0	0.9
7		Beeswax	6.0	6.0	0.9	6.0
Feel	-	,				
Spr	Spread					-

As is apparent from the results shown in Tables 25 and 26, the mascaras of the Examples have very good feel and good spread at the time of application because the amphoteric urethane resin and silicone polymer are used in combination.

To the contrary, the mascara of the Comparative Example has good feel because the amphoteric urethane resin is used, but is inferior in spread at the time of application.

#### 10 Make-up cosmetics (foundations)

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#### Examples 1i to 7i, Comparative Examples 1i

## (i) Preparation of pigment

The respective components shown in Tables 27 and 28 described hereinafter were mixed in the proportion shown in the same tables and then pulverized through a pulverizer to prepare a pigment.

# (ii) Preparation of water phase

Deionized water was heated to 70 degree C and bentonite was added to make swollen. Then, sodium carboxymethylcellulose dispersed previously in propylene glycol was dissolved by adding to the resulting solution. Triethanolamine, methylparaoxybenozate, and one selected from amphoteric urethane resin or a silicone polymer were added and dissolved thereto to prepare a water phase.

# (iii) Preparation of oil phase

The respective components shown in Tables 27 and 28 described hereinafter were mixed in the proportion shown in the same tables and then dissolved with heating to prepare an oil phase.

# (iv) Preparation of pigment dispersion

A pigment dispersion was prepared by adding the above pigment to the water phase with stirring, followed by passing through a colloid mill.

#### (v) Emulsification

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The above pigment dispersion and oil phase were heated to 75 degree C and 80 degree C, respectively, and the above oil phase was added to the pigment dispersion with stirring. After cooling the dispersion, a perfume was added at 45 degree C, and the stirring was continued up to the room temperature to prepare a foundation, respectively.

Using the foundations of the Examples and Comparative Example thus obtained, the respective characteristics were evaluated according to the criteria for make-up cosmetics. The results are summarized in Tables 27 and 28 described hereinafter

(Foundation)

(Parts)

63.3 3.0 0.5 q.s. 2.0 2.0 8.5 q.s. 0.3 0.2 0.5 4.0 q.s. \_ 8.0 4.0 ą.s 63.3 2.0 0.5 q.s. 0.2 8.5 q.s. 0.5 4.0 8.0 q.s. 0.2 3.0 <u>(</u>@ 7. q.s. 3 63.3 q s (2) 2.0 2.0 3.0 q.s. 0.5 0.3 0.2 0.5 2.4 8.5 4.0 8.0 q s q.s 0.2 7 4.0 7 Examples 63.3 2.4 2.0 2.0 30 8.5 q.s. 0.5 q s. 0.3 0.5 7: 4.0 q.s. 0.2 0.5 8.0 ą.s Sodium carboxymethylcellulose Propylene glycol monostearate Amphoteric urethane resin (a) Methyl paraoxybenzoate Propyl paraoxybenzoate Cetostearyl alcohol Isopropyl myristate Coloring pigment Silicone polymer Propylene glycol Triethanolamine Delonized water Liquid paraffin Titanium oxide Liquid lanolin Stearic acid Bentonite (Types) Talc Perfume Spread Feel Pigment Oil phase · Water phase

(Foundation)

		Examples	ş		Comparative Example
		5j	6)	7.j	1]
	Stearic acıd	2.4	2.4	2.4	2.4
	Propylene glycol monostearate	2.0	2.0	2.0	2.0
	Cetostearyl alcohol	0.2	0.2	0.2	0.2
	Liquid lanolin	2.0	2.0	2.0	2.0
əse	Liquid paraffin	3.0	3.0	3.0	3.0
bps	Isopropyl myristate	8.5	8.5	8.5	8.5
!!0	Propyl paraoxybenzoate	q.s.	q.s.	qs.	q.s.
	Amphoteric urethane resin (a)	0.5	0.5	0.5	0.5
	Silicone polymer	0.3	0.3	0.3	
	(Types)	(2)	. (9)	(2)	
	Deionized water	63.3	63.3	63.3	63.6
	Sodium carboxymethylcellulose	0.2	0.2	0.2	0.2
əs	Bentonite	0.5	0.5	0.5	0.5
eyd	Propylene glycol	4 0	4 0	4.0	4.0
1916	Triethanolamine	1.1	1.1	1.1	1.1
?M	Methyl paraoxybenzoate	q.s.	q.s.	q.s.	q.s.
	Titanium oxide	8.0	8.0	8.0	8.0
tnə	Talc	4.0	4.0	4.0	4.0
швід	Coloring pigment	q.s.	q.s.	q.s.	q.s.
Per	Perfume	q.s.	q.s.	q.s.	q.s.
Feel	70				
Spr	Spread				

As is apparent from the results shown in Tables 27 and 28, the foundations of the Examples have very good feel and good spread at the time of application because the amphoteric urethane resin and silicone polymer are used in combination.

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To the contrary, the foundation of the Comparative Example has good feel because the amphoteric urethane resin is used, but is inferior in spread at the time of application.

As described above, according to the cosmetics of the present invention, the amphoteric urethane resin and silicone polymer cause micro phase separation and the silicone polymer is unevenly distributed on the surface, thereby making it possible to provide the surface with the smoothness. As a result, the cosmetics are superior in spread at the time of application without impairing the touch when using the amphoteric urethane resin. The cosmetics of the present invention exhibit the excellent water resistance to neutral water as a result of ion bond(s) between the carboxyl group(s) and the tertiary amino group(s), while they exhibit excellent cleansing properties to shampoo as a result of the debonding of ions. Furthermore, the cationic tertiary amino group in the amphoteric urethane resin interacts with the surface of negatively charged hairs to exhibit good adhesion.

When using an aqueous solution or a water dispersion of a silicone polymer as the silicone polymer, the compatibility with the amphoteric urethane resin is enhanced to some degree, thereby making it possible to prepare a cosmetic having good stability.

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When structural units derived from ethylene oxide, as a nonionic hydrophilic component, is introduced into the structure of the amphoteric urethane resin, sufficient hydrophilicity is obtained and the hair washability are particularly improved when using as the hair cosmetic.

When a polysiloxane bond(s) is/are introduced into the structure of the above amphoteric urethane resin, the touch is particularly improved furthermore when using as the hair cosmetic.

### What is claimed is:

- A cosmetic comprising an amphoteric urethane
   resin having carboxyl group(s) and tertiary amino
   group(s) in a molecule, and silicone polymer.
- 2. The cosmetic according to claim 1, wherein said silicone polymer is a nonionic silicone polymer.
- 3. The cosmetic according to claim 1, wherein said silicone polymer is a polyether-modified silicone polymer.
- 4. The cosmetic according to claim 1, wherein said silicone polymer is a phenyl-modified silicone polymer.
- 5. The cosmetic according to claim 1, wherein said silicone polymer is an amino-modified silicone 20 polymer.
  - 6. The cosmetic according to claim 1, wherein said silicone polymer is an alkyl-modified silicone polymer.

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- 7. The cosmetic according to claim 1, wherein said silicone polymer is an alkoxy-modified silicone polymer.
- 5 8. The cosmetic according to claim 1, wherein said silicone polymer is a cyclic silicone polymer.
- 9. The cosmetic according to any one of claims 1 to 8, wherein said silicone polymer is in the form of an aqueous solution or a water dispersion.
- 10. The cosmetic according to any one of claims 1 to 9, wherein said amphoteric urethane resin has structural unit(s) derived from ethylene oxide in its 15 structure.
  - 11. The cosmetic according to any one of claims 1 to 9, wherein said amphoteric urethane resin has polysiloxane bond(s) in its structure.

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12. The cosmetic according to any one of claims 1to 11, wherein said amphoteric urethane resin is in the form of an aqueous solution.

13. The cosmetic according to any one of claims
1 to 12, which is at least one selected from the group
consisting of hair cosmetics, skin care cosmetics and
make-up cosmetics.

# ABSTRACT OF THE DISCLOSURE

The present invention provides a cosmetic which is superior in both characteristics of feel and spread at the time of application. The cosmetic is a cosmetic comprising an amphoteric urethane resin having carboxyl group(s) and tertiary amino group(s) in a molecule, and a silicone polymer.





# **DECLARATION FOR PATENT APPLICATION**

As a below named inventor, I hereby declare that:

My residence, post	office address and citizenship	are as stated below next to my nan	e.	
I believe that I am to of the subject matter the specification of	er which is claimed and for which	tor (if only one name is listed below th a patent is sought on the invention	/) or an original, first and joint inventor ( n entitled <u>COSMETICS</u>	of plural names are listed below)
(check one)	☑ is attached hereto			
•	☐ was filed on			as
	Application Serial No and was amended on			(if applicable).
I hereby state that referred to above.	I have reviewed and understar	nd the contents of the above identif	ed specification, including the claims, a	as amended by any amendment
I acknowledge the Regulations, §1.56		which is material to the patentab	ility of this application in accordance	with Title 37, Code of Federal
I hereby claim fore and have also iden claimed:	ign priority benefits under Title tifled below any foreign applica	35, United States Code, §119 of a ation for patent or inventor's certific	any foreign application(s) for patent or i ate having a filing date before that of th	nventor's certificate listed below e application on which priority is
Prior	Foreign Application(s)			Priority Claimed
	PCT/US00/21874	PCT	10 August 1999	<b>Yes</b> No
	(Number)	(Country)	(Day/Month/Year Filed)	res No
	(Number)	(Country)	(Day/Month/Year Filed)	Yes No
_	(Number)	(Country)	(Day/Month/Year Filed)	Yes No
the filing date of the	e prior application and the natio  (Application Serial No.)	nal or PCT international filing date of the control	tle 37, Code of Federal Regulations, §1 of this application:  (Status-patented, pendin	<del></del>
	(Application Serial No.)	(Filing Date)	(Status-patented, pendin	ng, abandoned)
	he following attorney(s) and/or		ition and to transact all business in th	e Patent and Trademark Office
connected herewith	n: No 26.863: Laurelee A. Dunc	can Red No. 44,096; Jane F. Ger	naro, Reg. No. 34,884; Karen G. Kaise	er. Reg. No. 33,506; Thomas F.
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true; and further t imprisonment, or b	hat these statements were m	ade with the knowledge that willi	If that all statements made on information of the like so not that such willful false statements made that such willful false statements made that such willful false statements made in the statements made on information in the statements are statements.	nade are punishable by fine or
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Declaration			Docket No. 1888P
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